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Aerosol optical properties calculated from size distribution measurements: An uncertainty study

Hagen Telg[a,b], Don R. Collins[c], and Allison McComiskey[d]

[a]Global Monitoring Laboratory, National Oceanic and Atmospheric Administration, Boulder, Colorado, USA; [b]Cooperative Institute for Research in Environmental Sciences, University of Colorado, Boulder, Colorado, USA; [c]Department of Chemical and Environmental Engineering, University of California, Riverside, California, USA; [d]Brookhaven National Laboratory, Upton, New York, USA

ABSTRACT

We use Monte Carlo uncertainty propagation to estimate the uncertainty of aerosol scattering coefficients, σ_s, that have been derived from measured particle size distributions. We consider the particular case where the size distributions are measured using a combination of a scanning mobility particle sizer (SMPS) and an aerodynamic particle sizer (APS). Uncertainties that are propagated include those intrinsic to the instruments and those that arise from variabilities in aerosol microphysical properties, including particle shape, density, and complex refractive index. Particular emphasis is put on the size dependent counting efficiency of both instruments which have weaknesses in a particle size range that dominates aerosol optical properties. The T-matrix method is utilized to simulate the effect of particle shapes on σ_s. To narrow the probability distribution of aerosol properties we discuss uncertainties for a single geographic location, which is the Southern Great Plains site (SGP) of the Department of Energy's Atmospheric Radiation Measurement (ARM) User Facility. We estimate a 95% confidence interval for σ_s between −40% and +68%. A partial dependence analysis, for which we use generalized additive models, identifies uncertainties in counting efficiency and particle shapes as the dominant contributors to the size of the confidence interval.

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1. Introduction

The simplicity with which aerosols are represented in climate models is the dominant cause for deviations between the modeled and the measured effect of aerosol on the Earth's radiation budget (Mann et al. 2014). Knowledge gaps in aerosol properties are a major contributor to the total uncertainty in the Earth radiation budget and the radiative forcing of climate as simulated in global climate models (Boucher et al. 2013). Pathways to reduce model uncertainty include the improvement of how aerosol microphysical and chemical properties such as particle size distribution and chemical classification are represented in the model (Mann et al. 2014) and how those properties are used to derive aerosol optical properties (Barnard et al. 2010). As climate models improve, it becomes increasingly important to have a comprehensive understanding of uncertainties in measured aerosol properties and the assumptions that we make to explain their relationships. Further, as we demand more from measurements to characterize the relationships among properties, the ability to perform high-quality closure studies also becomes increasingly important. While optical properties can be observed directly with good accuracy, e.g., by using a nephelometer, the ability to accurately estimate optical properties based on microphysical characteristics is essential to consider variations throughout the atmosphere caused by physical and chemical processes. In particular, the particle size distribution is the most fundamental property and influences the processes which bring about spatio-temporal distributions in aerosol optical properties. Among the instruments that are commonly used to measure size distributions of sub and super micron sized particles are the scanning mobility particle sizer (SMPS) and the aerodynamic particle sizer (APS), respectively. Closure studies, where directly measured aerosol optical properties are compared to those derived from microphysical properties, show large variations in agreement (Buonanno et al. 2009; Cai, Montague, and Deshler 2011; Quinn et al. 1996; Wex et al. 2002). Furthermore,

CONTACT Hagen Telg hagen.telg@noaa.gov Global Monitoring Laboratory, National Oceanic and Atmospheric Administration, 325 Broadway, Boulder, CO 80305, USA.

the observed deviations often approach or even exceed the boundaries of a suggested 99% confidence interval of ±20%, indicating that the interval is likely underestimated. To the best of our knowledge previous studies have not considered the effect of particle shapes and did not adequately take instrument uncertainties into account. Specifically, the SMPS and APS instruments have uncertainties that vary with particle size and are particularly large in diameter ranges relevant to aerosol optical properties.

In recent efforts to determine instrument uncertainties in atmospheric aerosol measurements, intercomparison studies were conducted between instruments of the same type; that is instruments that are based on the same measurement technique, but which were independently developed by different companies and research institutions. This included one intercomparison study of 7 SMPS and a second study of 15 APS instruments, where the prior study focuses on Aitken and accumulation mode particles ranging from 5 to 500 nm and the latter on predominantly coarse mode particles from 500 to 5000 nm (Pfeifer et al. 2016; Wiedensohler et al. 2012, 2018). Both studies show that for most parts of the measured particle number size distributions measured concentrations at a given diameter deviate by no more than ±10%. It has also been pointed out that for certain diameter ranges, deviations greatly surpass this value, in particular, for particles larger than 200 nm and smaller than 900 nm for the SMPS and APS, respectively (Pfeifer et al. 2016; Wiedensohler et al. 2012). Unfortunately, this particle size range is responsible for a large amount of scattered light in ambient aerosols. For the United States Southern Great Plains site (SGP) of the Department of Energy's Atmospheric Radiation Measurement (ARM) User Facility we estimate that, during the year of 2012, more than 60% of the scattered light comes from particles with diameters between 200 and 900 nm. Furthermore, uncertainties in derived aerosol optical properties due to uncertainties in particle shapes, in particular of accumulation mode particles, were largely neglected in previous studies (Buonanno et al. 2009; Cai, Montague, and Deshler 2011; Quinn et al. 1996; Wex et al. 2002). Although the understanding of the shapes of ambient particles is currently limited, there have been some tentative values derived from laboratory studies that can be utilized in an analysis of uncertainty (e.g., Alexander et al. 2016).

In this paper we present a study of uncertainties in the aerosol scattering coefficient, $\sigma_{s,}$ that is approximated from an aerosol particle size distribution, which is measured with a combination of a SMPS and an APS instrument. We estimate $\delta\sigma_{s,}$ the uncertainty of σ_{s}, by using a Monte Carlo method to propagate instrument specific uncertainties and uncertainties in aerosol properties, which affect measurements as well as calculations of optical properties. Instrument uncertainties taken into account include sizing accuracy and counting efficiency, while aerosol property uncertainties encompass variability in particle shapes, their densities and the complex refractive index. Aerosol properties have a certain characteristic for a geographic region. Also, most of the considered uncertainties depend to some extent on the particle size which means that a change in the shape of the particle size distribution results in a change in the resulting uncertainty. As a demonstration, we utilize the distinctive properties of aerosols and the mean size distribution obtained from ARM's SGP user facility for the year 2012, as illustrated in Figure 1.

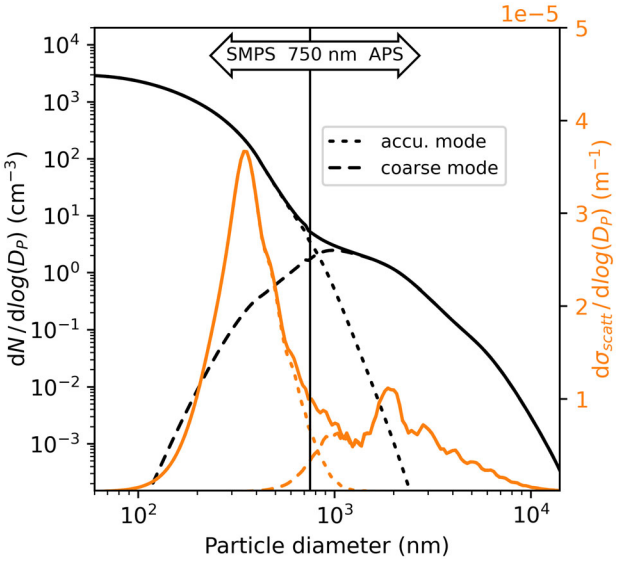

Figure 1. Black graph, left ordinate: average size resolved number concentration of dry aerosol particles for the year 2012 measured by a combination of SMPS and APS at SGP. Particle diameters represent mobility equivalent diameters below 750 nm and aerodynamic equivalent diameters above 750 nm with a narrow transition regime where the overlap of the two measurements are merged (for details see Collins 2010b). Gray (orange) graph, right ordinate: size resolved scattering coefficient derived from the size distribution for light with a wavelength of 550 nm and the assumption of spherical particles with a refractive index of 1.5+*i*0. Short and long dashed lines show contributions from accumulation and coarse mode particles, respectively, which were derived by fitting log-normal distributions to the size distribution.

2. Methods

In this section, we outline uncertainties that are being considered, the method employed to propagate them,

and the estimated quantities. Finally, we provide brief overviews of relevant measurement techniques and theoretical approaches.

2.1. Uncertainties and their origins

This study aims to determine the uncertainties that must be taken into account when calculating the aerosol scattering coefficient from a measured particle size distribution, where the size distribution represents a composite of measurements from an SMPS and an APS instrument. In addition to instrument-related intrinsic uncertainties, we consider those uncertainties arising from the variability of aerosol microphysical properties. Since those properties are rarely measured, the most common approach is to assume idealized particles of perfect sphericity and a particular and uniform chemical composition. Deviations and variability of ambient particles from those ideal particle properties result in uncertainties in the sizing of the particles as well as the calculated optical properties. In the following we discuss which aerosol properties are considered and how they influence measurements and models.

When a particle deviates in its properties from that of an ideal particle the diameter that is measured by the particle sizing instrument does not reflect the physical diameter of the particle but the equivalent diameter of the ideal particle that would result in the same measurement (Thomas and Charvet 2017). A SMPS instrument measures a particle's size based on its mobility and provides the mobility equivalent diameter d_m. The APS, according to its measurement technique, reports the aerodynamic equivalent diameter d_a. On the other hand, theoretical techniques to estimate optical properties of aspherical particles, like the T-matrix method which is applied here (see below), use the volume equivalent diameter d_V (Mishchenko 1991). When approximating aspherical particles as spheroids these diameters have the following relationship,

$$d_m = d_V \cdot \chi \tag{1}$$

$$d_a = d_V \cdot \sqrt{\frac{\rho_p}{\rho_0 \cdot \chi}} \tag{2}$$

where χ is the dynamic shape factor, ρ_0 and ρ_p are the densities of the calibration particles and the measured (ambient) particle, respectively. Note, in this study we calculate aerosol optical properties using the T-matrix method which describes particle shapes by the ratio of equatorial to polar radius ξ (Mishchenko 1991). Here we will quantify particle shapes with the shape parameter Φ, which is equal to $\xi - 1$ for oblate and $1 - 1/\xi$ for prolate spheroids. To convert between χ and Φ we use an empirical relationship which was established by Davies (1979). In addition to the size and shape the optical properties depend on the real and imaginary part of the complex refractive index $n + ik$ which changes due to variations in the particle's chemical composition and abundance of black carbon, respectively. Throughout this study, we assume aerosols to be dry with a controlled relative humidity below 40%. Higher relative humidities and the associated hygroscopic growth would significantly increase the complexity of the problem.

In conclusion of the preceding paragraph, with the assumptions and approximation applied here, we can estimate the overall uncertainties of calculated aerosol optical properties from size distribution based on the measurements intrinsic sizing and counting efficiency uncertainties, δd and δce, respectively, and uncertainties in the particles shape parameter Φ, the real n and imaginary part k of the complex refractive index, and the particle's gravimetric density ρ_p.

2.2. Monte Carlo method

A Monte Carlo uncertainty study is a method for quantifying the uncertainty in the output of a model by using repeated simulations with randomly generated inputs (Possolo and Iyer 2017). Input parameters of the model encompass all variables with uncertainties that are propagated, here Φ, n, k, and ρ_p, and random values generated with a probability distribution in accordance with the particular uncertainty. The frequency distribution of the model outputs, here σ_s, describes the uncertainty of that parameter. The Monte Carlo method has several benefits when compared to traditional uncertainty analysis, particularly in its ability to incorporate non-linear relationships between inputs and outputs and covariance between different inputs, both of which are highly pertinent to the current study. In the next two sections we first discuss the model and then for each parameter the probability distribution from which the random values are selected.

2.2.1. Model

In addition to the variables with uncertainties our model has one more input that is not randomized, which is the original size distribution. Also, we split the size distribution at 750 nm into two separate parts, which are then handled individually. This implies that all other input parameters are also generated

individually, with different distributions for the two regimes. We justify this separation using the following assumptions. Different aerosol modes, in particular accumulation and coarse mode, are predominantly of physically different origin, which implies that properties and their uncertainties are largely independent of each other. The same applies for the fundamentally different measurement techniques of the SMPS and APS, which respond differently to changes in particle properties, as discussed in the previous section. The example size distribution in Figure 1 shows that the upper and lower limits of the SMPS and APS instruments, respectively, coincide roughly with the diameter where accumulation and coarse mode contribute equally to the scattering coefficient. Note, the tail of the accumulation mode (short dashed orange line) that is larger than 750 nm accounts for 8% of the total scattering coefficient of the coarse mode (long dashed orange line) and is expected to have only a minor effect on the presented results. The same applies to the tail of the coarse mode that is smaller than 750 nm, which accounts for 2% of the total scattering from the accumulation mode.

The input parameters δd and δce are random correction values for the particular instruments intrinsic sizing and counting efficiency errors, respectively. According to multi-instrument studies the sizing uncertainty is proportional to the particle size (Pfeifer et al. 2016; Wiedensohler et al. 2012, 2018). The model, therefore, applies a percentage change to all particle diameters, given by δd. The counting efficiency strongly depends on the particle diameter (Pfeifer et al. 2016; Wiedensohler et al. 2012, 2018). Therefore, δce is a multiplicator of a size dependent standard deviation which we estimate from multi-instrument intercomparisons of SMPS instruments by Wiedensohler et al. (2012, 2018) and APS instruments by Pfeifer et al. (2016). Figure 2 shows the counting efficiency uncertainties and the effect of δce. For a detailed discussion on the origins of the observed measurement discrepancies we refer the reader to the cited publications. Subsequently we perform a conversion of the measured diameters d_m and d_a to volume equivalent diameters d_V with input values for particle shape parameter Φ and density ρ_p using Equations (1) and (2) and the empirical relationship between Φ and χ according to Davies (1979). Finally, we simulate the scattering coefficient σ_s, which is returned by the model, from the resulting size distribution using the T-matrix method with input values for the real and imaginary part of the complex refractive index, $n + ik$, and the particle shape parameter Φ (Leinonen 2014; Mishchenko 1991).

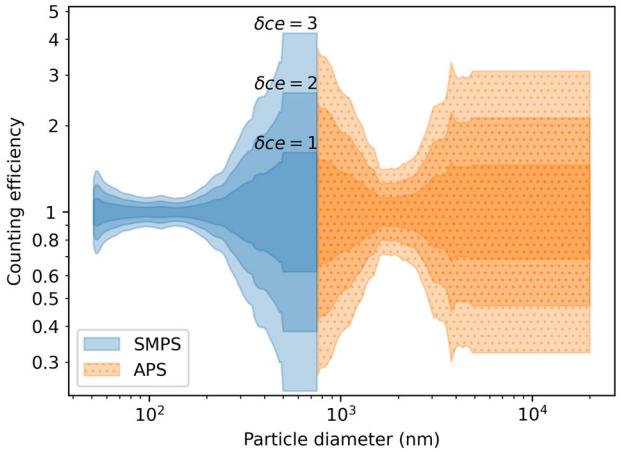

Figure 2. Diameter dependent counting efficiency for the SMPS (solid blue) and the APS (solid orange and hatch pattern of dots) instruments. Different shades indicate the standard deviations from the mean in multi-instrument intercomparison studies (Pfeifer et al. 2016; Wiedensohler et al. 2012). The model input δce is equivalent to the standard deviation.

2.2.2. Model inputs and their distribution

In the following we discuss the values which we applied in the Monte Carlo model. The short lifetime of aerosols causes significant spatial and temporal variability in their properties, leading to an excessively large uncertainty in calculated optical properties if all possible variations were taken into account. In this study we consider the particular conditions for the year 2012 at the Southern Great Plains site (SGP) of the Department of Energy's Atmospheric Radiation Measurement (ARM) User Facility. In 2012, many relevant observations were conducted simultaneously, and we are confident that a year worth of data represents a statistically relevant data set.

The size distribution that we use in our model, and which is shown in Figure 1, is the average size distribution that was measured by an SMPS and an APS instrument for the entire year (Collins 2010b, 2010a). Figure 3 displays the frequency distribution of input parameters used in the model for the accumulation mode section of the size distribution, as measured by the SMPS instrument for particles less than 750 nm in diameter. We use a normal distribution for the intrinsic sizing precision δd with a standard deviation of 2%, which is estimated from a multi-instrument intercomparison study by Wiedensohler et al. (2012). The counting efficiency parameter, as discussed above, scales the size dependent counting efficiency and is by definition normally distributed with a standard deviation of one. Only few studies exist that make detailed

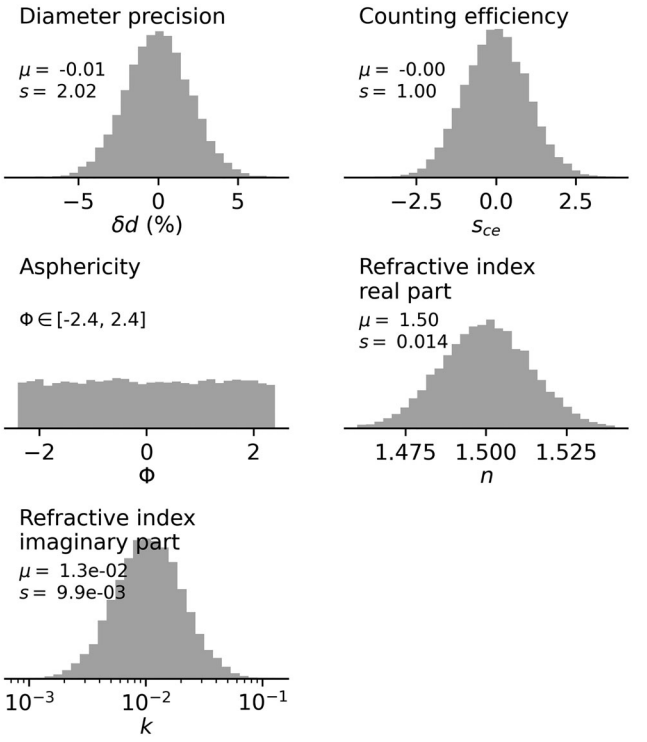

Figure 3. Frequency distributions of model input parameters for the diameter regime $d < 750$ nm, which is dominated by accumulation mode particles that are measured with the SMPS instrument. For normal and log-normal distributions the mean, μ, and standard deviations, s, are given.

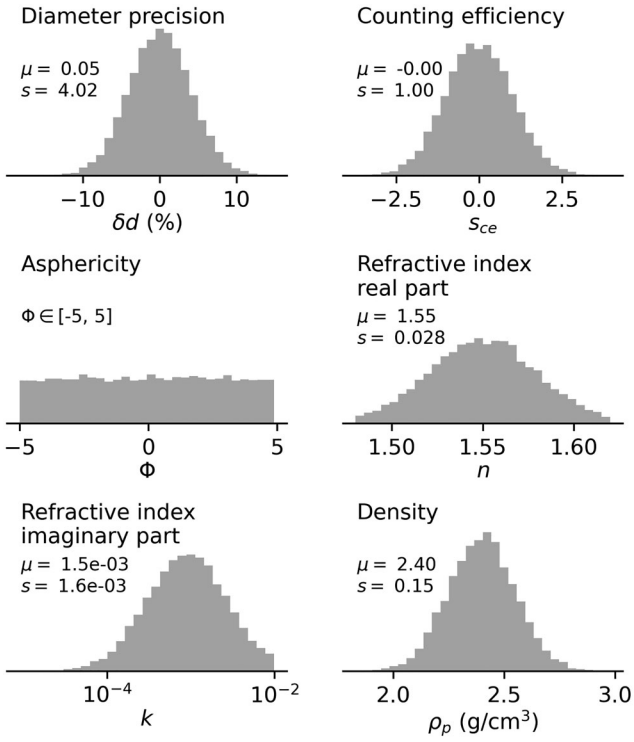

Figure 4. Frequency distributions of model input parameters for the diameter regime $d > 750$ nm, which is dominated by coarse mode particles that are measured with the APS instrument. For normal distributions the mean, μ, and standard deviations, s, are given.

observations of the shape of aerosol particles, and none were conducted at SGP during 2012. Here we assume a uniform distribution with $\Phi \in$ [-2.4, 2.4]. It has been demonstrated that the particle shape parameter of aerosols describes rather an even than a normal distribution (Alexander et al. 2015; Meland et al. 2010). The boundaries for Φ are the equivalent of an effective dynamic shape factor χ of 1.06, which was estimated for accumulation mode ammonium sulfate particles in a laboratory experiment (Zelenyuk, Cai, and Imre 2006). Like the shape, the complex refractive index is not well constrained and measurements in particular of the imaginary part carry a large uncertainty (e.g., Washenfelder et al. 2013). For the real part n we use the frequency distribution that was derived from measurements with an aerosol chemical speciation monitor (ACSM) during 2012 at SGP and which resembled a normal distribution with a mean of 1.50 and a standard deviation of 0.014 (see ACSM section for details). The technique that we use to derive n is not able to provide the imaginary part k of the refractive index. Also, none of the instruments that can be used to determine k, like an imaging polar nephelometer or cavity ring down spectrometer (e.g., Reed Espinosa et al. 2017; Washenfelder et al. 2013), were deployed at SGP during 2012. Therefore, we estimate a mean k of 0.013 and a standard deviation of 0.01 from the frequency distribution of absorption coefficients measured with an aethalometer for submicron particles (Sherman et al. 2015), the average size distribution, and Mie theory. Note, we use a log normal distribution for k due to the large ratio between the standard deviation to the mean and the requirement of k to be positive.

Figure 4 displays the frequency distribution of model input parameters for the coarse mode section of the size distribution, as measured by the APS instrument for particles more than 750 nm in diameter. We use a normal distribution for the intrinsic sizing precision with a standard deviation of 4%, which was determined in a multi-instrument intercomparison study by Pfeifer et al. (2016). The counting efficiency parameter, as discussed above, is normally distributed with a standard deviation of one. Coarse mode aerosols are known to contain particles with larger asphericities than accumulation mode particles. A large range of effective dynamic shape factors have been published, some of which result in unlikely Φ equivalents (Eidhammer, Montague, and Deshler 2008). Here we use $\Phi \in$ [-5, 5] in accordance with

recent observations by Alexander et al. (2015). Assuming quartz and feldspar as the predominant constituents of dust particles at SGP suggest a real part of the refractive index between 1.5 and 1.6. Similar values have been determined from dust particles in various studies at different locations around the world (Eidhammer, Montague, and Deshler 2008). Based on these two statements and a lack of precise measurements of n at SGP, we assume a normal distribution of n with a mean of 1.55 and a standard deviation of 0.03. Note, the ACSM instrument deployed at SGP cannot measure super-micron particles and can therefore not be used to determine n in coarse mode particles. Applying the same approximation as for the accumulation mode in the previous section we estimate a mean imaginary part k of the refractive index in super-micron particles of $1.4 \cdot 10^{-3}$ with a standard deviation of $2 \cdot 10^{-3}$. To convert the aerodynamic particle diameters that are measured by the APS instrument to the volume diameter the model additionally requires the particle's volumetric mass density ρ_p. We assume ρ_p to be normally distributed around $2.4\,g/cm^3$ with a standard deviation of $0.15\,g/cm^3$ based on values reported in the literature (Chen et al. 2011; Reid et al. 2003, 2008; Rocha-Lima et al. 2018). Note, this is slightly lower than what would be expected from quartz ($\sim 2.55\,g/cm^3$) and feldspar ($\sim 2.76\,g/cm^3$).

2.3. Scanning mobility particle sizer (SMPS)

The SMPS instrument measures aerosol particle sizes based on the particle's electrical mobility, which is determined by the amount a particle is deflected in an electric field (Collins 2010b; Knutson and Whitby 1975). Under well controlled conditions – constant flow rates, pressure, temperature, etc. – the amount a particle is deflected depends on its size and shape and the number of charges on the particle. Ambient aerosols carry an unknown number of charges which are adjusted to a bipolar charge distribution using α-radiation in a so-called neutralizer. In a SMPS only particles that are deflected by a particular amount are measured at the exit of the instrument using a condensation particle counter. In order to measure different electrical mobilities, thus particle sizes, the strength of the electric field is scanned across the required range. To derive the particle size distribution from the measured mobility distribution a sophisticated inversion routine is applied that takes an assumed charge distribution into account. An SMPS by itself is not able to determine effects from the particle shape, so reported diameters are those of spherical particles that have the equivalent electrical mobility. This measurement technique is commonly limited to particles in the sub-micron diameter range. For the instrument used here the upper diameter limit was 750 nm (Mahish and Collins 2017).

2.4. Aerodynamic particle sizer (APS)

The APS measures aerodynamic properties of particles (Baron 1986), in particular, the speed of each particle after it passes through the accelerated flow within a nozzle. For a given flow rate, temperature, and pressure the particle speed depends on its size, density, and shape. Similar to the SMPS the reported diameter is the equivalent for a spherical particle with a particular density that has the equivalent aerodynamic properties. Note that a commonly used calibration standard – spherical polystyrene beads – have a density close to $1.055\,g/cm^3$ (manufacturer information, Thermo Fisher). Since ambient aerosol particles typically have a higher density, it is common practice to normalize diameters to the equivalent of a higher density particle. At SGP for example, diameters are normalized to match those of particles with a density of $2\,g/cm^3$ (Collins 2010b). Most APS instruments are designed to measure super-micron particles. The lower diameter limit for the instrument used in this study is 500 nm.

2.5. Aerosol Chemical Speciation Monitor (ACSM)

The Aerosol Chemical Speciation Monitor (ACSM) is an instrument that can measure mass loading and chemical composition of aerosol particles (Ng et al. 2011). Particles that enter the instrument are vaporized on a 600 °C surface. Resulting fragments are then ionized with an electron beam and their mass analyzed with a quadrupole mass spectrometer. The ACSM distinguishes between organic, sulfate, nitrate, ammonium, and chloride. The particular instrument that was deployed at SGP in 2012 had an upper particle size detection limit of 1 μm (Watson 2017; Zawadowicz and Howie 2016). It has been demonstrated that the real part of the refractive index can be estimated based on electrolyte refractive indexes and their concentrations, which are derived from measured ion concentrations (Brock et al. 2015; Zaveri 2005).

2.6. T-matrix method

A common technique to calculate optical properties of particles of arbitrary shape is the transition matrix

(T-matrix) method (Waterman 1965). In the presented study we use a formulation of the T-matrix method that approximates particles as randomly oriented oblate and prolate spheroids (Leinonen 2014; Mishchenko 1991), where the asphericity is quantified by the ratio, ξ, of the equatorial to polar radius. The model further requires the particle's complex refractive index and the wavelength of the scattered light, where the latter is kept constant throughout this study at a value of 550 nm. As ξ increases, calculations become too costly to be carried out for every possible combination of random input parameters in the Monte Carlo analysis. Therefore, we compiled a four-dimensional look-up table (LUT) for the scattering coefficient as a function of particle diameter, shape parameter, real, and imaginary part of the refractive index. We linearly interpolate the LUT to the random input values. Note, the T-matrix code exhibits increasing difficulties in computing larger particle shape parameters with increasing particle diameter. For particles larger than 2 μm extrapolation was required for large Φ.

3. Results and discussion

Figure 5 shows the distribution of scattering coefficients σ_s that are produced when the Monte Carlo model is executed with the parameter distributions described above. Note, the abscissa is scaled logarithmically and σ_s has a log-normal distribution, which is appropriate for a strictly positive measure. When the entire size distribution is considered (Figure 5, top) we estimate a 95% uncertainty interval ($2s$) from -40% to $+$ 68%. By treating the accumulation mode ($d < 750$ nm) and coarse mode ($d > 750$ nm) independently, the standard deviation of the total σ_s is given by the square root of the sum of the squares of the standard deviations in the individual regimes. This implies even larger uncertainties when the two regimes are considered separately, with uncertainty intervals of [-51, +106] % and [-49, +96] %, respectively (see center and bottom of Figure 5).

Most if not all of the propagated uncertainties vary slowly in time. Aerosol microphysical properties are known to change on a time scale of days, and instrument drift happens on even longer time scales. Uncertainties will therefore manifest in a constant or slowly changing bias rather than high frequency noise. To investigate the contribution of each input parameter to the total uncertainty we perform multivariate analysis using generalized additive models, GAM, (Servén and Brummitt 2018; Wood 2017). To

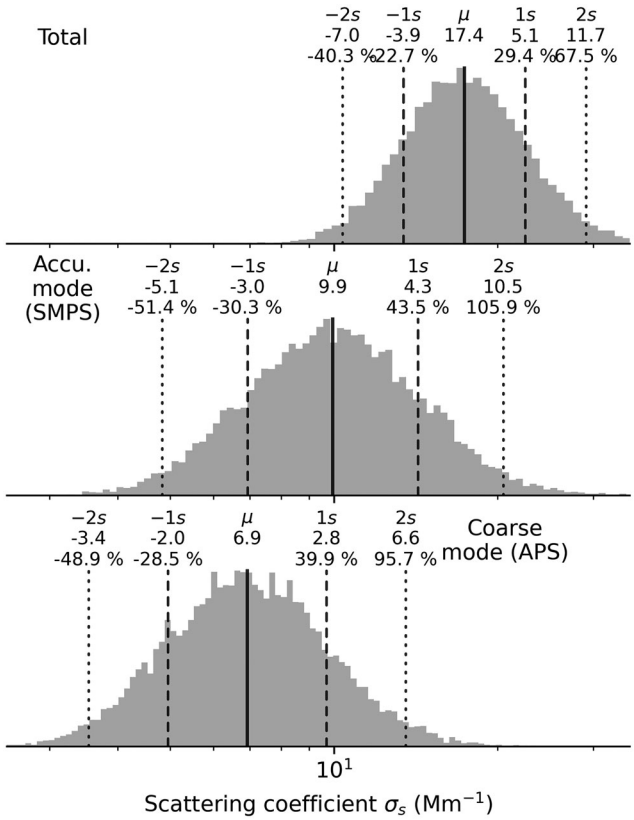

Figure 5. Frequency distributions of the computed scattering coefficients for the entire (top), the accumulation mode dominated part (center) and the coarse mode dominated part (bottom) of the size distribution. For each the mean, μ, and standard deviations, s, for a log-normal distribution are given.

demonstrate trends, we employ a basic model that considers all variables independently and disregards any interactions.

Figures 6 and 7 show the GAM analysis which we conduct separately for the two diameter regimes, $d < 750$ nm and $d > 750$ nm, respectively. The partial dependence function, which has been normalized to the mean scattering coefficient, is depicted by the solid black line in each plot for the respective variable. In addition, we show in blue the frequency distribution of the random parameters (shown in gray) projected onto the partial dependency function. This distribution and its standard deviations, illustrate how much the uncertainty of the respective parameter affects the calculated scattering coefficient. For both diameter regimes it stands out that uncertainties in the counting efficiencies are the leading source for uncertainty. It can be argued that careful calibration would reduce this uncertainty. However, the lack of a standard size distribution or a technique that measures the "true" size distribution hampers such efforts. The use of a third technique, e.g., optical particle

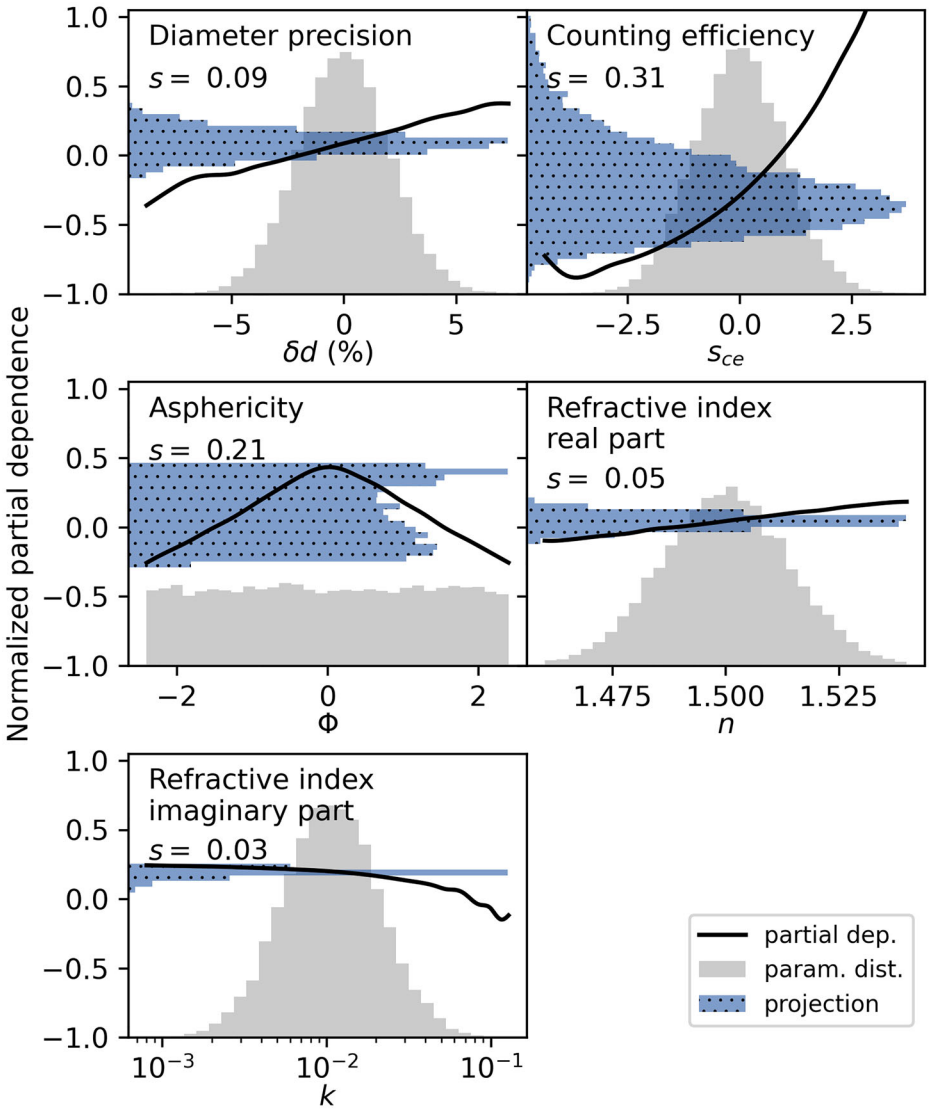

Figure 6. Black lines, normalized partial dependences of the scattering coefficient on the different input parameters for the accumulation mode dominated diameter regime. Histogram without hatching, parameter frequency distribution (same as in Figure 3). (Blue) histogram with dotted hatching, frequency distribution of input parameters after projection onto the respective partial dependency function.

sizing, might have the potential to improve this uncertainty. The second largest source for uncertainty originates from particle asphericities, which have three noteworthy characteristics that distinguish it from other parameters. First, even the relatively small particle shape parameters considered for accumulation mode particles (Figure 6) are a large source for uncertainty. Second, while all other parameters show a mostly monotonic partial dependence function the particle shape parameter is symmetric with respect to the ordinate at Φ equal 0. Third, the effect of asphericity on σ_s is opposite for the two diameter regimes. This implies that approximations of σ_s which assume spherical particles will result in a high bias in the $d < 750$ nm regime and a low bias in the $d > 750$ nm regime, when any aspheric particles are present. Contributions from the remaining parameters are significantly smaller. Therefore, only small improvement is expected in the overall σ_s uncertainties when those parameters are better constrained, e.g., through automated size calibrations, or improved retrievals of the complex refractive index and volumetric mass density through better measurements of aerosol particle's chemical compositions. Note, these partial dependence functions can be used to estimate how the σ_s uncertainty is affected when a parameter's probability distribution differs from that which is assumed here.

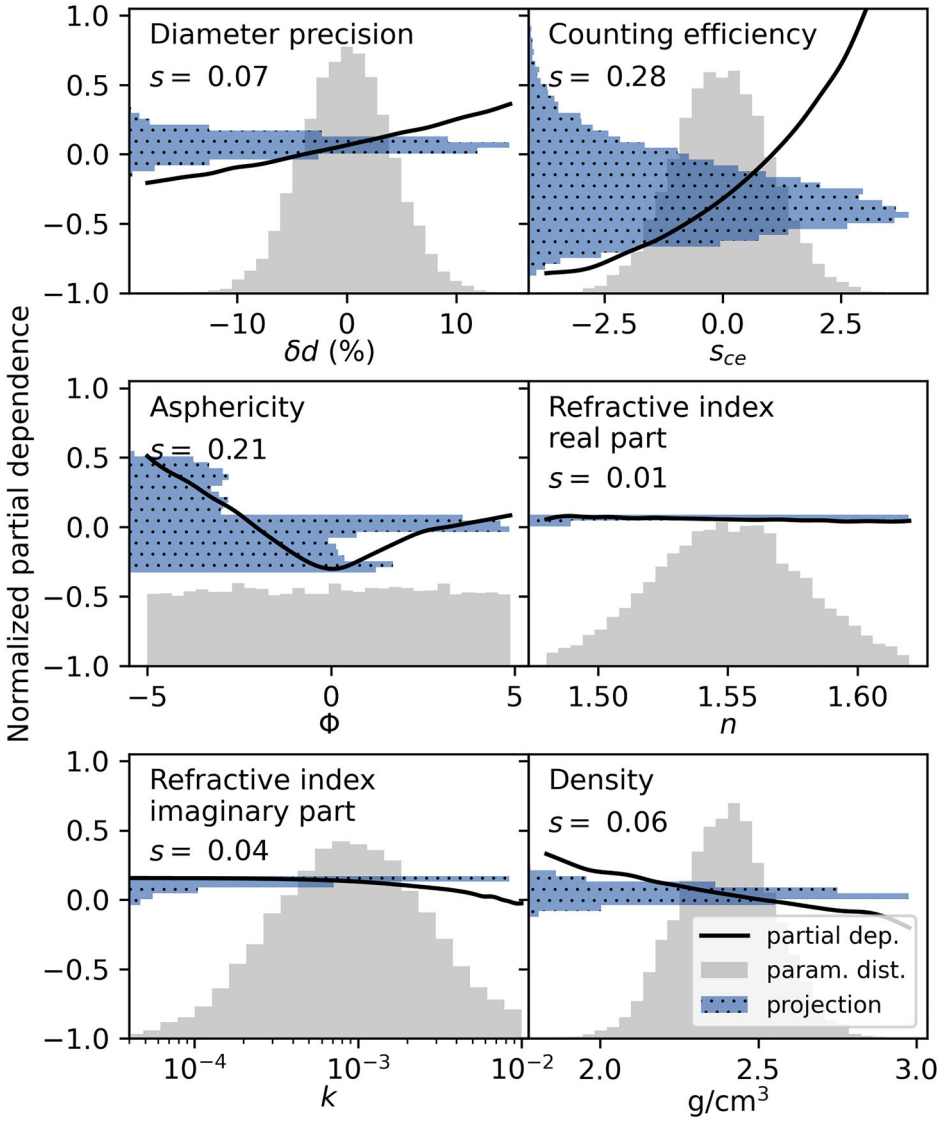

Figure 7. Same as Figure 6 but for the diameter regime that is dominated by coarse mode particles.

4. Conclusions

We presented an uncertainty study of aerosol scattering coefficients that have been derived from size distribution measurements with an SMPS and an APS instrument. Using a Monte Carlo method, we propagate uncertainties in the instrument's intrinsic sizing and counting efficiency, the shape of ambient aerosol particles, the aerosol particle complex refractive index, and the particle's volumetric mass density. We find a 95% confidence interval between −40% to + 68% for the entire particle size distribution. When accumulation mode (measured with the SMPS) and coarse mode (measured with the APS) are treated separately we find almost identical uncertainty intervals from roughly −50% to +100%. We find that the largest sources for uncertainty in the scattering coefficient originate from uncertainties in the counting efficiency in either instrument and in uncertainty in particle shapes. The presented study stands out from previous research as it considers uncertainties related to measurement techniques and microphysical properties of aerosols in more detail. However, some limitations remain as we continue to apply assumptions that considerably simplify the scenario, such as assuming a spheroid morphology and homogeneous composition of aerosol particles. Fully understanding the implications of those assumptions is difficult as conducting relevant simulations and observations presents a challenge.

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ORCID

Hagen Telg http://orcid.org/0000-0002-4911-2703
Don R. Collins http://orcid.org/0000-0002-0010-290X
Allison McComiskey http://orcid.org/0000-0002-6125-742X

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